

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

20-4576P

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/319202
NEW

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/JP97/04388

December 2, 1997

December 3, 1996

TITLE OF INVENTION

METHOD FOR PRODUCING MOLDED ARTICLE OF MODIFIED POLYTETRAFLUOROETHYLENE BY JOINING

APPLICANT(S) FOR DO/EO/US

SHIRASAKI, Osamu; KAWACHI, Shoji; TANIGAWA, Shingo

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(3)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(2)).
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.w/ International Search Report
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
 - 1). (3) Three Sheets of Formal Drawings

U.S. APPLICATION NO (if known, see 37 CFR 1.5)		INTERNATIONAL APPLICATION NO		ATTORNEY'S DOCKET NUMBER	
NEW		PCT/JP97/04388		20-4576P	

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5):
 Neither international preliminary examination fee (37 CFR 1.482)
 nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO
 and International Search Report not prepared by the EPO or JPO. **\$970.00**

International preliminary examination fee (37 CFR 1.482) not paid to
 USPTO but International Search Report prepared by the EPO or JPO **\$840.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO
 but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. **\$760.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO
 but all claims did not satisfy provisions of PCT Article 33(1)-(4) **\$670.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO
 and all claims satisfied provisions of PCT Article 33(1)-(4) **\$96.00**

ENTER APPROPRIATE BASIC FEE AMOUNT =

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30
 months from the earliest claimed priority date (37 CFR 1.492(e)).

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total Claims	4 - 20 =	0	X \$18.00
Independent Claims	1 - 3 =	0	X \$78.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			NO + \$260.00
TOTAL OF ABOVE CALCULATIONS =			\$ 840.00
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).			\$ 0.00
SUBTOTAL =			\$ 840.00
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			\$ 0.00
TOTAL NATIONAL FEE =			\$ 840.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +			\$ 40.00
TOTAL FEES ENCLOSED =			\$ 880.00

Amount to be:
refunded

\$

charged

\$

CALCULATIONS

PTO USE ONLY

a. ☒ A check in the amount of \$ **880.00** to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account. No. _____ in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 02-2448.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR
1.137(a) or (b)) must be filed and granted to restore the application to pending status.

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32,868
REGISTRATION NUMBER

20-4576P

IN THE U.S. PATENT AND TRADEMARK OFFICE

APPLICANT: Osamu SHIRASAKI et al.

INT'L. APPLN. NO.: PCT/JP97/04388

SERIAL NO.: NEW

GROUP:

FILED: June 2, 1999

EXAMINER:

FOR: METHOD FOR PRODUCING MOLDED ARTICLE OF MODIFIED
POLYTETRAFLUOROETHYLENE BY JOINING

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents
and Trademarks
BOX PATENT APPLICATION
Washington, D.C. 20231

June 2, 1999

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

IN THE SPECIFICATION:

Before line 1, insert --This application is the national phase under 35 U.S.C. §371 of prior PCT International Application No. PCT/JP97/04388 which has an International filing date of December 2, 1997 which designated the United States of America.--

IN THE CLAIMS:

Claim 3, line 1, delete "or 2"

Claim 4, line 1, change "any one of claims 1 to 3" to --claim 1--

R E M A R K S

The specification has been amended to provide a cross-reference to the previously filed International Application.

The amendment to the claims corrects any improper multiple dependent claims to place the application into better form.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. §1.16 or under 37 C.F.R. §1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By 

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(Rev. 11/16/98)

SPECIFICATION

METHOD FOR PRODUCING MOLDED ARTICLE OF MODIFIED

POLYTETRAFLUOROETHYLENE BY JOINING

FIELD OF THE INVENTION

5 The present invention relates to a method for producing a
molded article of a modified polytetrafluoroethylene (hereinafter
referred to as "modified PTFE") by joining. In particular, the
present invention relates to a method for producing the molded
article of a modified PTFE by sintering the premolded parts of
10 the modified PTFE to join them together at their abutting faces.

PRIOR ART

A method is known to produce the molded article of a modified
PTFE, which comprises heating and press-bonding two or more molded
parts of a modified PTFE (see Hostaflon Information 21 (August,
15 1983) published by Hoechst AG). However, this method has
drawbacks such that a material loss is large since parts are
produced by cutting, and that faces to be bonded should be
processed so as to be smooth. Furthermore, in this method, it
is difficult to control pressure during press-bonding, and thus
20 defective products tend to be produced due to too high or low
pressures.

JP-B-63-67808 discloses the bonded product of the molded
articles of a modified PTFE. However, the articles are bonded
by heating and press-bonding. That is, the articles are bonded
25 by heating while applying external pressure. This method may be
suitable for bonding thin articles such as sheets or films, but
tends to produce defective products due to too high or low
pressures when thick articles are bonded by this method, like in

the case of the above method described in the brochure of Hoechst AG.

JP-A-55-57429 discloses a method comprising inserting a center mold in a tubular mold, placing cap molds at the top and bottom of the tubular mold, and bonding the both edges of a PTFE sheet in a space formed by the molds with expansion pressure of the sheet. This method requires a set of premolding molds and molds for expansion bonding for each size of a product, and is industrially disadvantageous as a method for producing various kinds of products. Furthermore, the molds should be sealed since the method utilizes the expansion pressure. Therefore, the steps of the method become very complicated as a whole.

Furthermore, when a thick wall container having a bottom and a sidewall is produced, a method is known, which comprises filling a modified PTFE powder in a mold and pressing it while heating. However, the compression distance is short at the bottom, while the distance is long at the sidewall. Thus, cracks may be formed at parts near the interface between the bottom and the sidewall, so that good molded articles are not produced.

SUMMARY OF THE INVENTION

One object of the present invention is to provide a method for producing the molded article of a modified PTFE by joining, in which bonded parts have sufficient bond strength even in the case of a thick wall article, and which overcomes the above-described drawbacks of the prior art.

Another object of the present invention is to provide a method for producing the molded article of a modified PTFE by joining, which can suppress the production of defective articles,

improve the dimensional accuracy of the article, and decrease the material loss due to the production of parts by cutting.

A further object of the present invention is to provide a method for producing the molded article of a modified PTFE by joining, which includes simplified operations to join the parts of a modified PTFE, and greatly improves the productivity, particularly in the mass production.

Accordingly, the present invention provides a method for producing the molded article of a modified PTFE by joining comprising the steps of:

providing at least two premolded parts of modified PTFEs having different coefficients of thermal shrinkage with allowing their joining faces to be in contact each other or to be closely placed, and

sintering the parts to join them at the joining faces.

In one preferred embodiment, the joining of the parts at their joining faces is carried out without the application of an external pressure.

In general, a premolded part has a coefficient of thermal shrinkage in the range between 0.2 and 10 %, and the difference of the coefficient of thermal shrinkage is from 0.2 to 9.8 % between two premolded parts which are adjacently placed.

In other preferred embodiment, at least one premolded part is surrounded by another premolded part having a larger coefficient of thermal shrinkage than that of at least one premolded part.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a graph showing the relationship of a premolding

pressure and a coefficient of shrinkage of the outer diameter of tubular premolded parts of the modified PTFEs having different particles sizes, which are used in Example 1.

Fig. 2 is a graph showing the relationship of a premolding pressure and a coefficient of shrinkage of the outer diameter of the tubular premolded parts of modified PTFE, which is used in Example 2.

Fig. 3 is a graph showing the relationship of a premolding pressure and a coefficient of shrinkage of the outer diameter of the premolded parts of modified PTFEs having different particles sizes, which are used in Example 3.

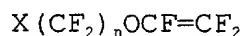
Fig. 4 is a graph showing the relationship of a premolding pressure and a coefficient of shrinkage of the outer diameter of the premolded parts of modified PTFEs having different particles sizes, which are used in Example 4.

Fig. 5 is a graph showing the relationship of a premolding pressure and an outer-diameter shrinkage factor of the premolded parts of modified PTFEs having different particles sizes, which are used in Example 5.

DETAILED DESCRIPTION OF THE INVENTION

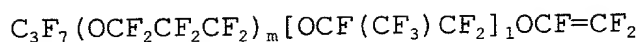
The term "modified PTFE" intends to mean a copolymer of tetrafluoroethylene and a small amount of one or more of the following modifier monomers.

Examples of the modifier monomers for PTFE include fluorine-containing unsaturated monomers other than TFE, such as a fluoroalkyl vinyl ether of the formula:



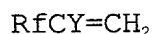
wherein X is a hydrogen atom, a fluorine atom or a chlorine atom,

and n is an integer of 1 to 6, or the formula:



wherein m and 1 are each an integer of 0 to 4, provided that they are not 0 (zero) at the same time;

- 5 $\text{CF}_3-\text{CF}=\text{CF}_2$, $\text{CF}_2=\text{CFH}$, $\text{CF}_2=\text{CFCl}$, $\text{CF}_2=\text{CH}_2$; an unsaturated compound of the formula:



wherein Rf is a linear or branched polyfluoroalkyl group having 3 to 21 carbon atoms, and Y is a hydrogen atom or a fluorine atom;
10 and the like.

Such a modifier monomer is usually used in an amount of 0.01 to 1.0 wt. % of the weight of TFE.

In the present invention, modified PTFE can be used in the form of an as-polymerized powder, or in the form of granules which
15 are obtained by granulating such a powder. Herein, the powders and granules of modified PTFE are collectively called "modified PTFE powder".

Furthermore, it is possible to disperse suitable amounts of various fillers in a modified PTFE powder. Examples of such
20 fillers are glass fiber, graphite fiber, carbon fiber, bronze powder, etc. In addition, other conventional additives may be compounded in a modified PTFE powder.

The method of the present invention is particularly useful to produce a molded article in which one part (first part)
25 surrounds the other part (second part), for example, a container having a bottom and a sidewall surrounding the bottom, an article having a solid or hollow cylindrical body and a flange surrounding such a body, etc.

In the method of the present invention, the parts of modified PTFE can be joined without the application of external pressure. However, the supplemental application of external pressure is not excluded from the present invention.

5 According to the production method of the present invention, in the case of the above-described article in which one part (first part) surrounds the other part (second part), when the second part has a smaller coefficient of thermal shrinkage than that of the first part, the first part shrinks in a greater degree than the
10 second part in the sintering step (heating and subsequent cooling) in the production method, so that a sufficient force may be exerted at their joining faces, and thus the parts can be bonded at their joining faces.

 The premolded parts of modified PTFE used in the method of
15 the present invention may be film-form, sheet-form, block-form or any other form premolded parts, which are produced by compression molding, or any other suitable molding methods. The shapes of premolded parts may be suitably selected according to the shapes of final molded articles.

20 Conditions for the production of the premolded parts of modified PTFE are selected from the same ranges as those of the conventional methods. That is, a temperature is less than the melting point of modified PTFE, a pressure is one such that modified PTFE powder particles are well adhered at a given
25 temperature to form an integral premolded part.

 Two or more premolded parts of modified PTFE having different coefficients of thermal shrinkage can be obtained by various methods.

Typical methods are explained. For example, parts having different coefficients of thermal shrinkage can be obtained by separately premolding two or more modified PTFE powders having the same molecular weight but different particle sizes under the same conditions. In general, a premolded part has a smaller coefficient of thermal shrinkage, as a particle size of a modified PTFE powder is made larger.

Two ore more premolded parts having different coefficients of thermal shrinkage can be obtained by premolding the same modified PTFE powder under different pressures. In general, a premolded part has a smaller coefficient of thermal shrinkage, as a pressure is higher.

Alternatively, premolded parts of modified PTFE having different coefficient of thermal shrinkage can be obtained under the same premolding conditions from an as-polymerized modified PTFE powder, and a granulated product of such a modified PTFE powder. In general, a premolded part made from the granulated product has a smaller coefficient of thermal shrinkage than one made from the ungranulated modified PTFE powder.

Herein, "a coefficient of thermal shrinkage" (%) is a coefficient of thermal shrinkage of an outer diameter of a disk-form molded article having a thickness of 7 mm molded with a mold having an inner diameter of 65 mm, after sintering. The coefficient of thermal shrinkage of an outer diameter is calculated according to the following formula:

Coefficient of thermal shrinkage (%) =

$$\left[\frac{(\text{inner diameter of mold (65 mm)}) - (\text{outer diameter of sintered disk (mm)})}{(\text{inner diameter of mold (65 mm)})} \right] \times 100$$

A disk-form molded article is produced by supplying 50 g of modified PTFE powder in a mold, increasing a pressure at a rate of 25 - 30 mm/min. at room temperature, and pressurizing the powder under 300 kgf/cm² for 5 minutes, and sintered by heating the disk up to 360°C at a heating rate of 50°C/hr., maintaining the disk at 360°C for 6 hours, and then cooling to 100°C at a cooling rate of 50°C/hr.

The size of the disk-form article is measured after keeping it at room temperature for 8 hour or longer.

10 In the present invention, the coefficient of thermal shrinkage of a premolded part of modified PTFE is preferably in the range between 2 % and 10 %, more preferably in the range between 0.5 % and 2.0 %.

15 The difference of the coefficient of thermal shrinkage of a pair of adjacently placed premolded parts of modified PTFE is preferably in the range between 0.2 % and 9.8 %, more preferably in the range between 0.4 % and 1.0 %.

Premolded parts can be bonded by providing them with allowing their joining faces to be in contact each other or to be closely placed, and sintering them.

20 Herein, the phrase "allowing joining faces to be closely placed" means that the joining faces are separated at a distance at which the joining faces can be bonded in the sintering step by the difference of a coefficient of thermal shrinkage between premolded parts.

25 The upper limit of a sintering temperature is not limited. However, too high temperature causes the thermal decomposition of modified PRFE. Thus, a sintering temperature is less than the

thermal decomposition temperature of modified PTFE. Heating and cooling rates can be suitably adjusted by conventional methods.

A heating time is not limited. Premolded parts are heated for a period of time sufficient for shrinking the parts to bond
5 the joining faces of the parts with sufficient strength.

According to the bonding method of the present invention, the joining faces of parts can be strongly bonded even when the parts have a large thickness. Thus, the production of defective articles is decreased, the dimensional accuracy of a molded
10 article increases, and material loss due to the production of parts by cutting can be decreased.

The method of the present invention improves the productivity, particularly in mass production, since the production of articles can be repeated by very simple procedures
15 once the combination of two or more modified PTFE powders having different coefficients of thermal shrinkage is selected, and then specific molding conditions suitable for such a selected combination are fixed, when the molded articles of modified PTFE having a certain shape and certain sizes are produced.

20 EXCAMPLES

The present invention will be illustrated by the following examples.

Example 1

1) NEW POLYFLON M-112 (modified PTFE available from Daikin
25 Industries, Ltd.) was ground and sieved to obtain two kinds of modified PTFE powders having an average particle size of 28 μm and 55 μm , respectively. Average particle sizes were measured with a laser beam diffraction type (dry type) particle size

distribution analyzer (manufactured by SYNPADEC).

Each powder was compression molded at 25°C under a pressure of 150, 200, 250, 300 and 350 kg/cm² to obtain a tubular premolded part having an outer diameter of 100 mm, an inner diameter of 50 mm and a height of 50 mm.

Each tubular article was heated at 360°C to sinter the modified PTFE. The tubular article shrunk in the radial direction. Fig. 1 shows a graph plotting a coefficient of thermal shrinkage of an outer diameter against a molding pressure.

10 It can be understood from these results that an article produced from a modified PTFE having a smaller particle size has a larger coefficient of thermal shrinkage than one produced from a modified PTFE powder having a larger particle size, at any premolding pressure.

15 2) The modified PTFE powder having an average particle size of 28 µm was molded under a pressure of 300 kg/cm² to obtain a tubular premolded part having an outer diameter of 100 mm, an inner diameter of 50 mm and a height of 70 mm.

Separately, the modified PTFE powder having an average particle size of 55 µm was molded under the same pressure as above to obtain a disk-form premolded part having a diameter of 50 mm and a thickness of 15 mm.

The disk-form premolded part was inserted in the lowest position of the hollow part of the tubular premolded part to provide a bottom, and the composite of the premolded parts was heated at 360°C for 6 hours without applying any external pressure around the periphery of the tubular article.

After cooling, the tubular article and disk-form article,

which constituted the sidewall and bottom of the finished container, respectively, were firmly bonded, and the joined faces were watertight and airtight.

Example 2

- 5 1) NEW POLYFLON M-137 (modified PTFE available from Daikin Industries, Ltd. having an average particle size of 425 μm (measured according to ASTM D4894) was molded at 25°C under various pressures in a range between 100 and 500 kg/cm^2 to obtain a tubular premolded part having an outer diameter of 100 mm, an inner
10 diameter of 50 mm and a height of 50 mm.

Each tubular article was heated and sintered at a temperature of from 340 to 390°C. The tubular article shrunk in the radial direction. Fig. 2 shows a graph plotting a coefficient of thermal shrinkage of an outer diameter against a molding
15 pressure.

It can be understood from these results that an article molded under a higher premolding pressure has a smaller coefficient of thermal shrinkage when the particle size is the same.

- 20 2) The modified PTFE powder (NEW POLYFLON M-137) was molded under a pressure of 250 kg/cm^2 to obtain a tubular premolded part having an outer diameter of 100 mm, an inner diameter of 50 mm and a height of 70 mm.

Separately, the same modified PTFE powder as that used in
25 the above was molded under a pressure of 400 kg/cm^2 to obtain a disk-form premolded part having a diameter of 50 mm and a thickness of 15 mm.

The disk-form premolded part was inserted in the lowest

position of the hollow part of the tubular premolded part to provide a bottom, and the composite of the premolded parts was heated at a temperature of 340 to 390°C for 6 hours without applying any external pressure around the periphery of the tubular article.

5 The tubular article and disk-form article, which constituted the sidewall and bottom of the finished container, respectively, were firmly bonded, and the joined faces were watertight and airtight.

Example 3

10 1) NEW POLYFLON M-111 (modified PTFE available from Daikin Industries, Ltd.) was ground and sieved to obtain two kinds of modified PTFE powders having an average particle size of 31 μm (Powder A) and 55 μm (Powder B), respectively. An average particle size was measured with a laser beam diffraction type (dry type)
15 particle size distribution analyzer (manufactured by SYNPADEC).

Each powder (50 g) was charged in a mold having an outer diameter of 95 mm, an inner diameter of 65 mm and a height of 60 mm, compressed at 25°C at a compression rate of 25 to 30 mm/min., and maintained under a pressure of 200, 300 and 400 kg/cm² for
20 5 minutes to obtain a disk-form premolded part.

Then, the premolded part was heated from room temperature to 360°C at a heating rate of 50°C/hr., maintained at 360°C for 6 hours, and cooled to 100°C at a cooling rate of 50°C/hr. After that, the premolded part was maintained at room temperature for
25 8 hours or longer. The outer diameter of the disk was measured, and the coefficient of shrinkage of an outer diameter was calculated according to the above formula. The results are plotted in Fig. 3.

When a molding pressure is the same, the smaller particle size gives a larger coefficient of shrinkage of an outer diameter. When a particle size is the same, the higher molding pressure gives a smaller coefficient of shrinkage of an outer diameter.

- 5 2) Powder B was molded under a pressure of 300 kg/cm² to obtain a tubular premolded part having an outer diameter of 110 mm, an inner diameter of 80 mm and a height of 100 mm.

Separately, Powder A was molded under a pressure of 300 kg/cm² to obtain a disk-form premolded part having a diameter of
10 80 mm and a thickness of 10 mm.

The disk-form premolded part was inserted in the lowest position of the hollow part of the tubular premolded part to provide a bottom, and the composite of the premolded parts was heated from room temperature to 360°C at a heating rate of 50°C/hr.,
15 maintained at 360°C for 8 hours, and then cooled to 100°C at a cooling rate of 50°C/hr., during which no external pressure was applied around the periphery of the tubular article. After this heating treatment, the composite molded article was maintained at a temperature of 23 to 25°C for 8 hours.

- 20 3) Water or acetone was poured in the hollow space of the article to check liquid-tightness. No leakage of water or acetone was observed.

A circular film having a thickness of 250 μm was skived from the bottom of the molded composite article, and a rectangular
25 sample having a width of 10 mm and a length of 30 mm was cut out from each of four areas, which were on four directions extending orthogonally each other from the center of the circular film and crossed the bonded line. The tensile properties of each sample

were measured with a strength-elongation measuring instrument (AGS 500D manufactured by Shimadzu Corporation) at a pulling rate of 100 mm/min. A tensile strength was from 152 to 189 kgf/cm², and an elongation at break was from 280 to 330 %. The sample was
5 broken at an air-chucked part, and no breakage was observed at the bonded part.

Example 4

1) Powders A and B obtained in Example 3 were granulated to obtain Granulated Powders C and D each having an average
10 particle size of 650 μ m, which was measured according to ASTM D4894.

Each Granulated Powder (50 g) was molded under the same conditions as those in Example 3 to obtain a disk-form article, and the coefficient of shrinkage of an outer diameter was measured
15 in the same manner as that used in Example 3. The results are shown in Fig. 4.

When a molding pressure is the same, a smaller particle size gives a larger coefficient of shrinkage of an outer diameter. When a particle size is the same, a higher molding pressure gives a
20 smaller coefficient of shrinkage of an outer diameter.

2) A tubular premolded part having an outer diameter of 110 mm, an inner diameter of 80 mm and a height of 100 mm was produced by the same method as that in the step 2) of Example 3 except that Granulated Powder D was used in place of Powder B.

25 Separately, a disk-form premolded part having a diameter of 80 mm and a thickness of 10 mm was produced by the same method as that in the step 2) of Example 3 except that Granulated Powder C was used in place of Powder A.

The disk-form premolded part was inserted in the lowest position of the hollow part of the tubular premolded part to provide a bottom, and the composite of the premolded parts was heated from room temperature to 360°C at a heating rate of 50°C/hr., maintained at 360°C for 8 hours, and then cooled to 100°C at a cooling rate of 50°C/hr., during which no external pressure was applied around the periphery of the tubular article. After this heating treatment, the composite molded article was maintained at a temperature of 23 to 25°C for 8 hours.

3) Water or acetone was poured in the hollow space of the article to check liquid-tightness. No leakage of water or acetone was observed.

Four rectangular samples were cut out from the bottom of the molded article in the same way as that in the step 3) of Example 3, and the tensile properties of each sample were measured. A tensile strength was from 155 to 157 kgf/cm², and an elongation at break was from 210 to 220 %. The sample was broken at an air-chucked part, and no breakage was observed at the bonded part.

Example 5

1) NEW POLYFLON M-111 (modified PTFE available from Daikin Industries, Ltd. having an average particle size of 33 μm (measured with a laser beam diffraction type (dry type) particle size distribution analyzer)) (85 wt. parts) and glass fiber (manufactured by NIPPON SHEET GLASS CO., LTD.) (15 wt. parts) were mixed (Powder E).

Powder E was granulated to obtain a granulated powder having an average particle size of 650 μm (Granulated Powder F). The average particle size of the granulated powder was measured

according to ASTM D4894.

Each powder (50 g) was molded under the same condition as those in Example 3 to obtain a disk-form article, and its coefficient of shrinkage of an outer diameter was measured in the same way as that in Example 3. The results are shown in Fig. 5. When a molding pressure is the same, a smaller particle size gives a larger coefficient of shrinkage of an outer diameter, even if glass fiber is added to a powder. When a particle size is the same, a higher molding pressure gives a smaller coefficient of shrinkage of an outer diameter.

2) A tubular premolded part having an outer diameter of 110 mm, an inner diameter of 80 mm and a height of 100 mm was produced by the same method as that in the step 2) of Example 3 except that Granulated Powder F was used in place of Powder B, and the molding pressure was changed to 400 kg/cm².

Separately, a disk-form premolded part having a diameter of 80 mm and a thickness of 10 mm was produced by the same method as that in the step 2) of Example 3 except that Granulated Powder E was used in place of Powder A, and the molding pressure was changed to 400 kg/cm².

The disk-form premolded part was inserted in the lowest position of the hollow part of the tubular premolded part to provide a bottom, and the composite of the premolded parts was heated from room temperature to 360°C at a heating rate of 50°C/hr., maintained at 360°C for 8 hours, and then cooled to 100°C at a cooling rate of 50°C/hr., during which no external pressure was applied around the periphery of the tubular article. After this heating treatment, the composite molded article was maintained

at a temperature of 23 to 25°C for 8 hours.

3) Water or acetone was poured in the hollow space of the article to check liquid-tightness. No leakage of water or acetone was observed.

What is claimed is:

1. A method for producing the molded article of a modified polytetrafluoroethylene by joining comprising the steps of:

providing at least two premolded parts of modified
5 polytetrafluoroethylenes having different coefficients of thermal shrinkage with allowing their joining faces to be in contact each other or to be closely placed, and

sintering the parts to join them at the joining faces.

2. A method according to claim 1, wherein the joining of
10 said parts at the joining faces is carried out without the application of an external pressure.

3. A method according to claim 1 or 2, wherein said premolded part has a coefficient of thermal shrinkage in the range between 0.2 and 10 %, and the difference of the coefficient of thermal
15 shrinkage is from 0.2 to 9.8 % between two premolded parts which are adjacently placed.

4. A method according to any one of claims 1 to 3, wherein at least one premolded part is surrounded by another premolded part having a larger coefficient of thermal shrinkage than that
20 of at least one premolded part.

ABSTRACT

A method for producing a molded article of a modified polytetrafluoroethylene by joining having the steps of providing at least two premolded parts of modified polytetrafluoro-

5 ethylenes having different coefficients of thermal shrinkage with allowing their joining faces to be in contact each other or to be closely placed, and sintering the parts to join them at the joining faces. According to this method, the joining faces of parts can be strongly bonded even when the parts have a large

10 thickness. Thus, the production of defective articles is decreased, the dimensional accuracy of a molded article increases, and material loss due to the production of parts by cutting can be decreased. This method improves the productivity, particularly in the mass production, since the joining procedures

15 are simple.

Fig. 1

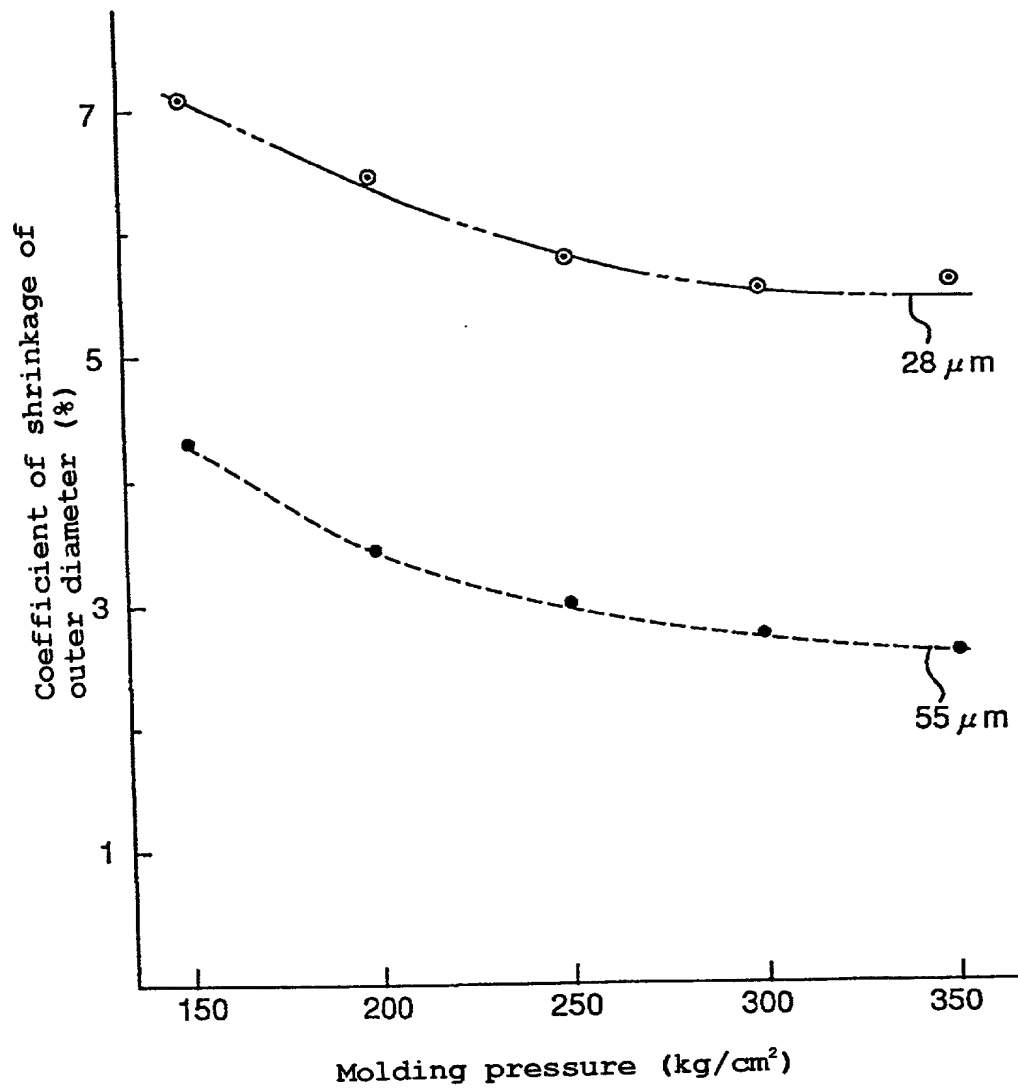


Fig. 2

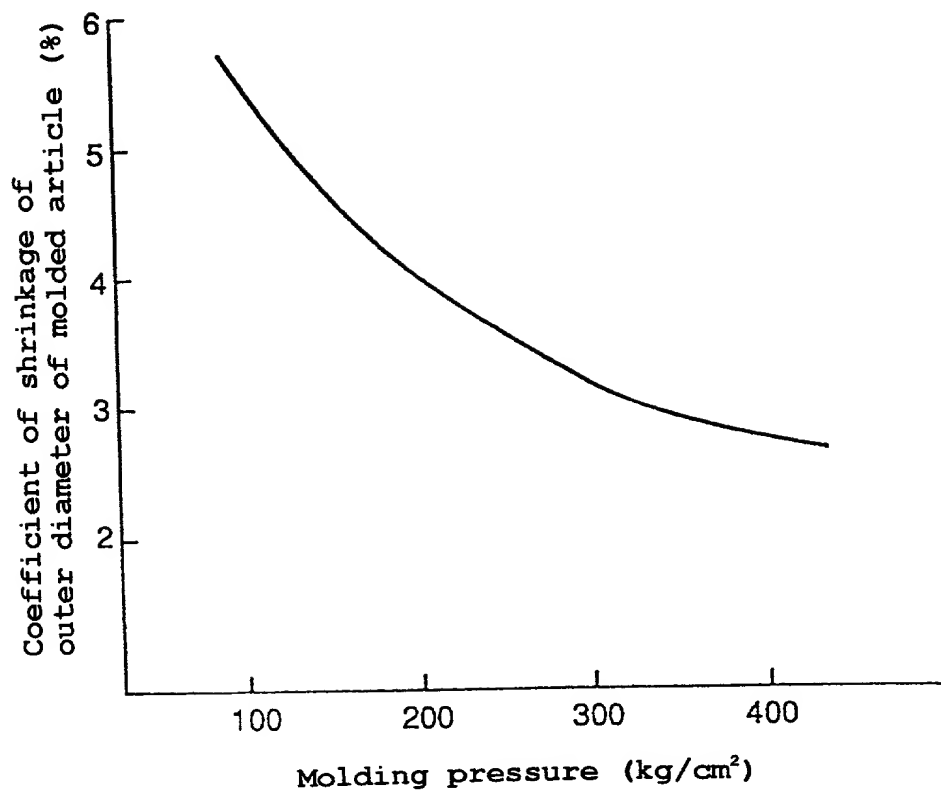


Fig. 3

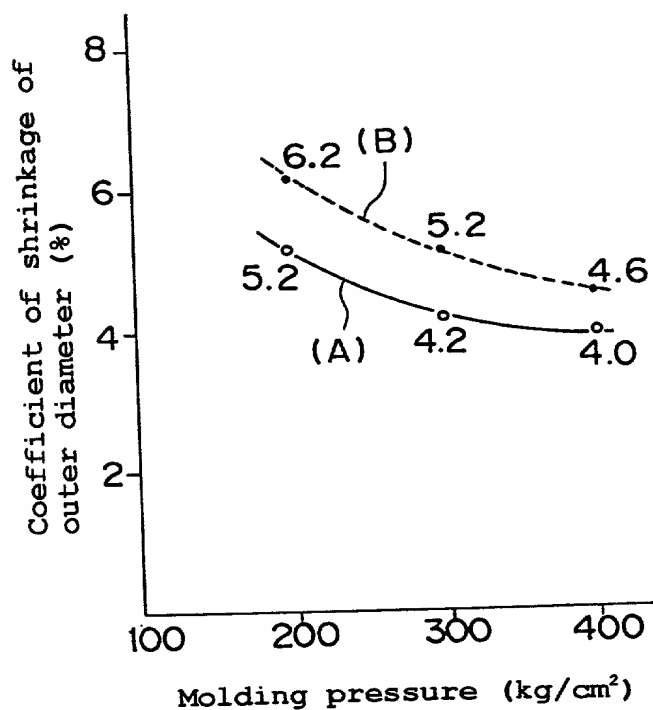


Fig. 4

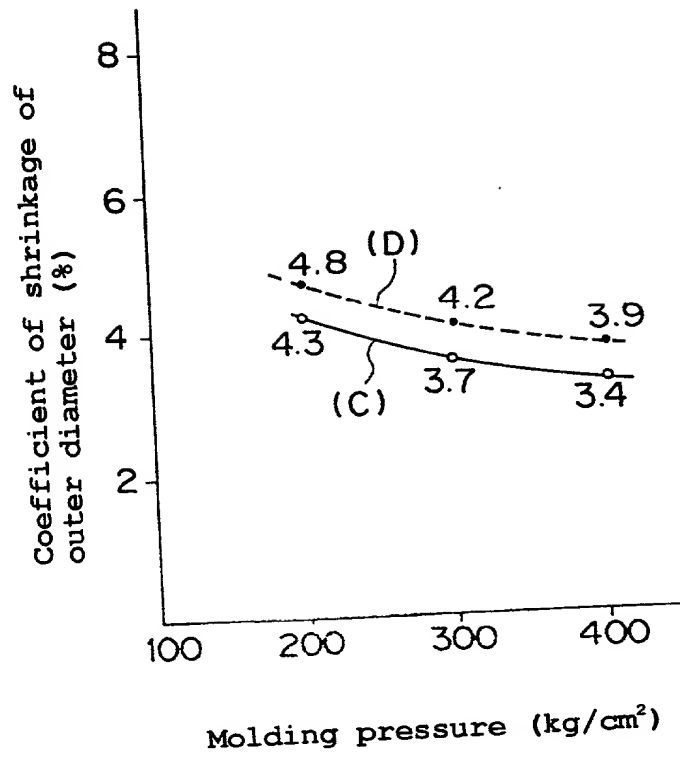
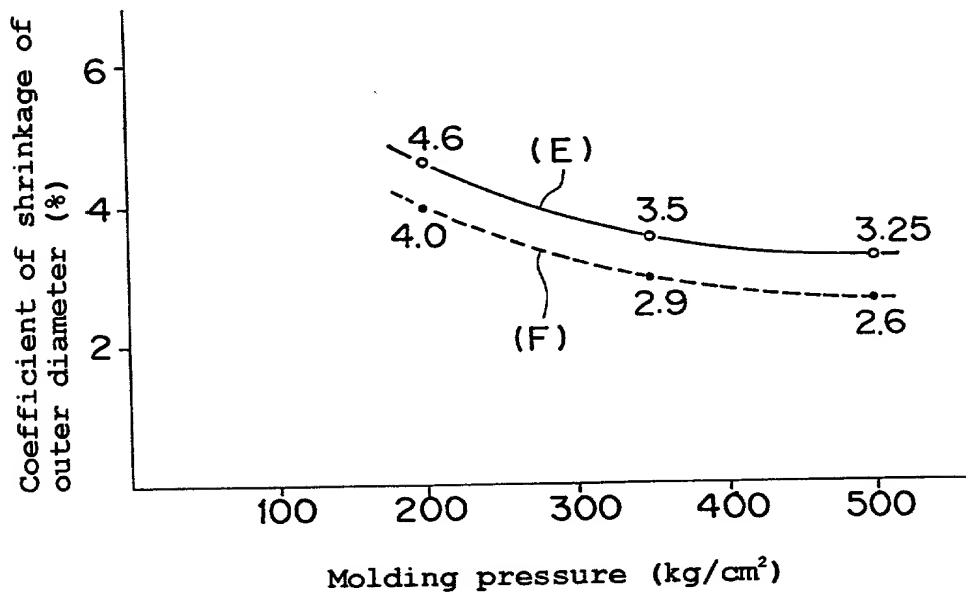


Fig. 5



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COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT AND DESIGN APPLICATIONS

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated next to my name; that I verily believe that I am the original, first and sole inventor (if only one inventor is named below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Insert Title: →

METHOD FOR PRODUCING MOLDED ARTICLE OF MODIFIED

POLYTETRAFLUOROETHYLENE BY JOINING

Fill in Appropriate
Information —

For Use →

Without

Specification

Attached:

the specification of which is attached hereto. If not attached hereto,

the specification was filed on _____ as
United States Application Number _____;
and amended on _____ (if applicable); and/or
the specification was filed on December 2, 1997 as PCT
International Application Number PCT/JP97/04388; and was
amended under PCT Article 19 on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe the same was ever known or used in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application, that the same was not in public use or on sale in the United States of America more than one year prior to this application, that the invention has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months (six months for designs) prior to this application, and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application by me or my legal representatives or assigns, except as follows.

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Prior Foreign Application(s)

Priority Claimed

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Information: →
(if appropriate)

<u>322544/1996</u> (Number)	<u>Japan</u> (Country)	<u>12/03/1996</u> (Month / Day / Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Month / Day / Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Month / Day / Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
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I hereby appoint the following attorneys to prosecute this application and/or an international application based on this application and to transact all business in the Patent and Trademark Office connected therewith and in connection with the resulting patent based on instructions received from the entity who first sent the application papers to the attorneys identified below, unless the inventor(s) or assignee provides said attorneys with a written notice to the contrary:

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